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Copper deposits in south-west England identified as a source of Copper Age metalwork

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Introduction

Large numbers of Bronze Age metal artefacts have been studied over the years with the aim of tracing their geological sources in order to comment on patterns of procurement and distribution, but the results have often been inconclusive (Budd et al. 1996). Here, we examine a very rare case study in which a combination of trace element and extremely individual lead isotope ratio data make it possible to provenance a small amount of archaeological metalwork. We show that a small, but important, group of Copper Age artefacts from the British Isles can only have been produced using copper from specific, highly unusual mineral deposits and that these are most probably sited within the Cornubian metallogenic province of south-west England.

The samples and their analysis

This paper is concerned specifically with five typologically related Copper Age artefacts, collectively described as group IMP-LI 2 (Rohl and Needham 1998, 87-8) on the grounds of their related lead isotope and trace element composition. They are described in (Table 1). Lead isotope analyses were undertaken by Rohl (1995) using a VG54 Thermal Ionisation Mass Spectrometer. ^{206}Pb/ ^{204}Pb errors are reported as 0.15% and ^{207}Pb/ ^{204}Pb errors as 0.2% (Rohl, pers comm.). Full analytical details are given by Rohl (1995). Analytical blanks were not reported.

Artefact	Description	Find site
WG2060	Halberd	Harbyrnig, Cumbria, UK
1905.11.6.3	Halberd	Maryport, Cumbria, UK
1927.7 - 13.1	Flat Axe	Old Shoyswell, Sussex, UK
84.83 H/1	Halberd	Castell Coch, South Glamorgan, Wales
84.83 H/3	Dagger	Castell Coch, South Glamorgan, Wales

Table 1. Description of the five LI-IMP 2 artefacts.

The artefacts have unusual $^{206}Pb/^{204}Pb$ ratios rising to over 41 (Table 2). Such high values are caused by the presence and radioactive decay of uranium in the ore source. As both ^{206}Pb and ^{207}Pb are derived from the decay of uranium isotopes (^{238}U and ^{235}U respectively), they will be found in a fixed ratio in samples taken from a single uranogenic ore body especially if that orebody is a low tonnage/high grade deposit. Such samples may plot in a linear array in $^{206}Pb/^{204}Pb$ vs. $^{207}Pb/^{204}Pb$ space with a gradient and intercept reflecting a geological event which is, in all probability, the age of mineralization (Faure 1986).

Artefact	$^{206}Pb/^{204}Pb$	$^{207}Pb/^{204}Pb$	$^{208}Pb/^{204}Pb$
WG2060	20.191	15.802	38.944
1905.11.6.3	33.429	16.472	38.663
1927.7 - 13.1	23.784	15.966	38.597
84.83 H/1	41.285	16.867	40.083
84.83 H/3	40.253	16.787	38.484

Table 2. Lead isotope ratios of the five LI-IMP 2 artefacts.

In this case, the five artefacts form a highly correlated linear array in $^{206}Pb/^{204}Pb$ vs. $^{207}Pb/^{204}Pb$ space. The artefacts lie on a Pb-Pb isochron with a model age of 205± 72Ma (95% confidence limits) based on the growth curve of Cumming and Richards (1975) (Figure 1). Correlated ^{204}Pb errors (r=0.998) were used in calculation. The relatively large error is a consequence of the small sample size (n=5).

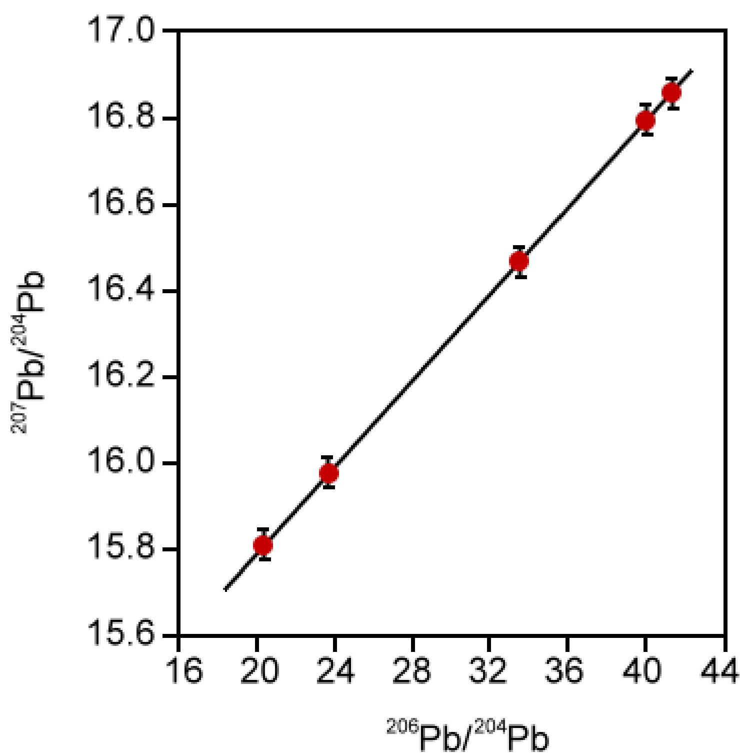

Figure 1. Lead isotope ratios of the five LI-IMP 2 artefacts (Rohl 1995, Rohl and Needham 1998). Data are plotted with 2σ errors reflecting reproducibility of measurements of the NBS981 standard as given by Rohl (1995) (note that $^{206}Pb/^{204}Pb$ errors are smaller than the symbols on this scale). The linear regression (M.S.W.D. of 0.972) gives a Pb-Pb isochron yielding a model age of 205$\pm$72Ma (2σ). The relatively large error is a consequence of the small sample size (n=5).

The shared isotopic characteristics and impurity pattern of the Copper Age IMP-LI 2 artefacts are so similar as to suggest that they were derived from the same mineral source. However, it is their highly unusual uranogenic lead isotope ratios and close fit to a single isochron (apparent on the ^{204}Pb plots used here) which make this almost certain; providing uniquely specific provenancing information. The lead isotope and compositional data indicate that the Copper Age IMP-LI 2 artefacts are highly likely to have been made from a U-, As-, Ni-bearing Cu ore, indeed, an ore where the original uranium content far exceeded that of the original lead - the mineralization was lead-poor. The data also show that the ore has a model age in the range 133-277Ma, namely early Permian to end Jurassic but probably of Triassic age. These dates coincide with the emplacement of late Variscan granites and associated post-Variscan mineralization events in Europe.

Potential ore sources

Numerous Bronze Age copper mines have now been identified in Britain and Ireland, but only one, Ross Island in Co. Kerry, has produced Copper Age dates (O'Brien, 1995) or contains exploitable arsenic-bearing (tennantite) ores (Ixer and Pattrick 2003; Ixer and Budd 1998). It is quite possible that this mine could have been the source of at least some Copper Age artefacts (Ixer in Prep.), but despite extensive sampling and investigation over the last ten years no U-bearing Cu mineralisation has been reported (Ixer and Pattrick 1995a, 1995b; 2003) and the ores contain only less-uranogenic lead (B. Barreiro pers. comm.). Of the Bronze Age mine sites investigated to date, only Great Orme near Llandudno in north Wales has Cu mineralisation containing uranogenic lead (Ixer and Budd 1998). The deposit also has a model age of 256±23Ma (Parnell and Swainbank 1990), within the range suggested for the IMP-LI 2 artefacts. However, detailed mineralogical investigations (Ixer and Stanley 1996, Ixer and Davies 1996) show that all the Great Orme copper ores are very As- and Ni-poor. Indeed "lead, zinc, cobalt and nickel minerals are absent" from the primary chalcopyrite-dolomite or secondary malachite-calcite ores from the Great Orme Mines (Ixer and Stanley, 1996).

The geochemistry of the five IMP-LI 2 artefacts is consistent with their being smelted using five metal association ores (Ag-Co-Ni-As-Bi ± U) which have a very restricted occurrence world-wide (Bastin 1993, Ixer 1990). In Europe the most famous and almost the only examples are the post-Variscan ores from Jachymov/Joachimstal in the Erzegeberge of Saxony, southern Germany (Baumann, 1976).

In Britain, a classical five metal association has been described from Tynebottom Mine in the North Pennines (Ixer and Stanley, 1987) and figured as such in Ixer (1990) based on material from the Natural History Museum collections, but the provenance of this material is now seriously compromised. There are undisputed five metal association deposits at Alva and Hilderston, in southern Scotland but these are too old, having a late Carboniferous age of 264-299Ma. They are also geochemically mismatched to the IMP-LI 2 metal as Hilderston carries very little copper mineralisation and at Alva nickel arsenides are very subordinate to cobalt arsenides (Hall *et al*. 1982, Stephenson *et al*. 1983). Alva and related and nearby deposits at the Bridge of Allan are also isotropically mismatched with $^{206}Pb/^{204}Pb$ ratios of only 18.134 - 18.485 (Parnell and Swainbank, 1984). These non-radiogenic values are confirmed by the petrography which shows uranium to be present only as a trace component of the ore (Parnell and Swainbank, 1984).

The <u>only</u> copper deposits in the British Isles, or adjacent areas of continental north-western Europe, which would have been accessible in antiquity and which meet the strict criteria dictated by their geological age and the compositional and lead isotope data would be those associated with the emplacement of granitic rocks of the Variscan orogeny. These granites mainly crop out within a series of isolated 'massifs', themselves within a west-east-striking belt across Europe from Cornwall and Devon (Cornubia) in the west, through Brittany (Armorica), the Massif Central and Vosges Mountains, to the Harz Mountains and the Erzegeberge in the east. Although five metal association ores from the latter two massifs could well match the IMP-LI 2 copper, other Continental ores from Variscan outcrops closer to the British Isles could not.

Artefact	**Cu**	**Sn**	**As**	**Sb**	**Ag**	**Pb**	**Fe**	**Ni**	**Co**	**Zn**	**Au**	**Bi**	**S**
WG2060	98.66	0.01	1.17	tr	0.05	0.01	0.02	0.02	tr	nd	0.05	tr	0.01
1905.11.6.3	95.70	0.01	3.77	0.10	0.04	0.03	0.20	0.07	tr	tr	0.02	0.06	0.01
1927.7 - 13.1	97	nd	1.4	0.04	0.02	0.01	0.03	0.09	nd	nd	na	0.02	na
84.83 H/1	96.7	nd	3.07	nd	0.05	nd	nd	0.16	nd	nd	nd	0.02	na
84.83 H/3	94.8	nd	4.59	tr	0.02	tr	0.01	0.50	nd	nd	0.02	0.01	na

Table 3. Chemical compositions of the five LI-IMP 2 artefacts (wt.%).

In the western part of the Variscan, namely the Cornubian granite batholith in SW England there are, albeit very rare, vein deposits that potentially have matching chemistries with the IMP-LI 2 copper (Bastin 1993). Some of these host a distinctive As-, Ni-, U-bearing mineralisation associated with Co, Bi and minor Ag (and Au) of the five metals type. It maybe relevant in this context that the uranogenic metalwork contains measurable Ag and Bi as well as As and Ni, although Co levels are considerably lower than Ni (Table 3). Some of the best known five metal association ores occur in two specific areas: around St. Stephen in Brannel, on the southern flank of the St. Austell granite (where mines have been worked in modern times for Cu, Ni and As as well as U (Dines 1956) and is close to the site of a gold-bearing placer) and at Wheal Bray on Bodmin Moor (C. Stanley pers.comm). These deposits are also of the correct age since the Cornubian mineralisation is a manifestation of the later stages of the Variscan orogeny; they are post-granite emplacement which is dated at 290-270Ma (Alderton, 1993).

Conclusion

When correctly presented and interpreted in the light of detailed mineralogical data from firmly contexted material, the combined lead isotope and compositional data show unambiguously that the five Copper Age IMP-LI 2 metal artefacts reported by Rohl (1995) were made from copper from a five metal association ore within a single source region in northwest Europe, namely Cornubia and perhaps, even within this region, the St. Austell and Bodmin Moor area. This is the first time it has been possible to provenance prehistoric metal artefacts with this degree of certainty and the highly unusual nature of the copper mineralisation associated with the five metals means that this is not a situation which is likely to occur often. Although it is clear from the archaeological evidence at Ross Island that copper was being produced there in the late third millennium BC from primary ores and not from secondary ores (O'Brien 1995, Ixer and Budd 1998) it is now apparent that Ireland was not the only source of Copper Age metal and that As-rich copper was also being made in Cornwall.

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